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Observations on Metallurgical Bonding Between Plasma Sprayed Tungsten and Hot Tungsten Substrates

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ABSTRACT. The effect of preheating metallurgically polished tungsten substrates on the bonding of plasma sprayed tungsten was investigated. The preheated temperatures of the substrate ranged from 2050 to 2700 F and the spraying was performed in an inert chamber using a nitrogen atmosphere. When the substrate temperature ranged between 2400 and 2700 F, a metallurgical bond was obtained between the coating and the substrate. At the higher substrate temperature (2700 F), the tungsten substrate recrystallized. However, when the preheating temperature was 2400 F, the substrate showed no signs of recrystallization. Electron micrographs showed that the initial coating-substrate interface was almost completely eliminated. This appears to have occurred by each substrate surface grain growing into the coating. *ASM-SLA Classification: L29, Q10c, 2-62; W.*

Author

WHILE CONSIDERABLE work has been carried out on plasma sprayed coatings, very little effort has been directed toward a critical examination of the resultant particle-to-substrate bonds. Since the characteristics of the initial bond generally determine the adherence and strength of bonding of any applied coating, improved bonding from one of mechanical interlock to one that is truly metallurgical is important. Therefore, an investigation was undertaken to explore the type and quality of coating-to-substrate bonds which can be obtained during plasma spraying. This work is a portion of a larger effort to characterize more fully the plasma-spray deposition phenomenon and to develop improved plasma-spray coatings.

In general practice, powder of one material is plasma sprayed on the roughened surface of a different material. In this investigation, however, tungsten powder was deposited on metallurgically polished tungsten substrates in order to minimize some of the variables involved in coating-substrate bonding. Admittedly, using the same material for coating and substrate and using highly polished substrate surfaces do not simulate general coating practice. However, these conditions were chosen so that the bond could be studied without undue complications. In this way, physical property variables such as thermal conductivity, linear coefficient of thermal expansion, relative melting temperatures, etc., were eliminated. Also, eliminating surface roughness excluded the questions of total roughness, roughness variation, and as-roughened surface composition. In addition, the smooth surface allowed additional deductions to be made on particle properties, such as plasticity, and permitted more controlled observations of effects on the substrate.

A preliminary investigation (unpublished) showed that plasma-sprayed tung-

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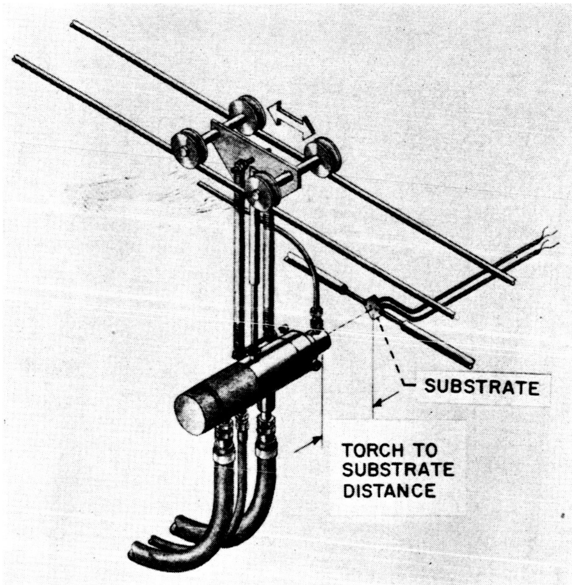

FIG. 1. Apparatus in inert chamber.

sten particles did not adhere to metallurgically polished tungsten specimens that were not preheated. However, tungsten particles did adhere to substrates of lower thermal conductivity, which reduced the quench rate of the particles and allowed them to cool at a slower rate. Therefore, this investigation was conducted on tungsten substrates that were preheated to relatively high temperatures, in order to reduce the quench rate of the particles and enhance their adherence.

To gain the maximum information from this investigation, the number of particles deposited on the substrate, in each test, was kept at a minimum. This was done to observe the characteristics of the individually sprayed particles rather than the characteristics of a gross deposit.

APPARATUS AND PROCEDURE

Commercial tungsten powder, 74μ to 30μ particle size, was sprayed on metallurgically polished tungsten coupons, 1 by 1 by 0.017-in. thick. These coupons were ground from as-received tungsten sheet, mounted in a cold castable mounting material, and metallurgically polished. After polishing, the specimens were broken out of their respective mounts and stored in a desiccator to prevent surface contamination. The particles were sprayed on the 1 by 1-in. surface that was metallurgically polished. The coupon thickness was kept to a minimum in order to permit rapid heat transfer to the back side for temperature monitoring.

All runs were made with a Thermal Dynamics F-40 torch operating in an environmental chamber (utilizing nitrogen) under the following fixed conditions: Number 1 nozzle—$^7/_{32}$-in. ID, 450 amp, 60 v, 80 SCFH* high purity dry nitrogen (plasma gas), 10 SCFH hydrogen (plasma gas), 10 SCFH high purity dry nitrogen (carrier gas), and, 11.9 setting on Continental Coatings hopper.

Each tungsten coupon was mounted by fitting two diagonally opposite specimen

* Standard cubic feet per hr.

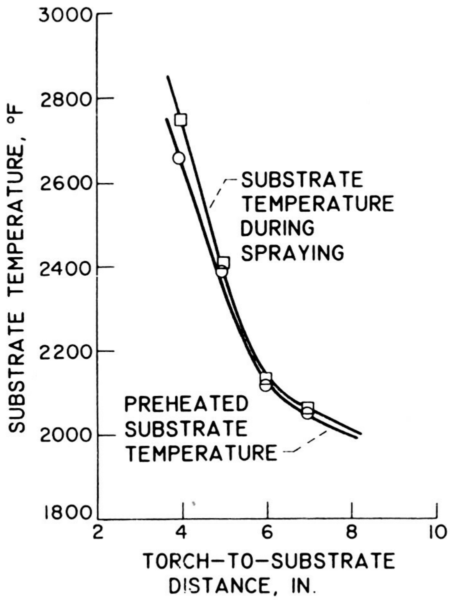

Fig. 2. Variation of substrate temperature with torch-to-substrate distance.

corners into spring-loaded alumina tubes. Each specimen had a Pt–Pt—13%–Rh thermocouple spot-welded to the back face and the temperature was monitored through the preheat and spray cycle by means of a recorder with a response of a few milliseconds. A schematic of the spray setup is shown in Fig. 1. The test sequence was as follows:

(*a*) Set coupon to torch distance
(*b*) Evacuate chamber to 20μ and backfill to atmospheric pressure with high-purity dry nitrogen
(*c*) Start torch and bring to operating conditions
(*d*) Roll torch into position and permit plasma gas to preheat tungsten coupon
(*e*) When specimen temperature reaches equilibrium (2 to 3 sec), activate powder spray for approximately 1 sec
(*f*) Roll torch out of position and shut down

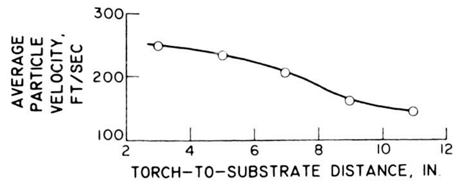

Fig. 3. Variation of average particle velocity with torch-to-substrate distance.

(*g*) Permit specimen to cool to room temperature and open tank.

The total time involved for specimen preheating, spraying, and cool down to 400 F was approximately 30 sec.

Average particle velocity measurements were made at various torch-to-substrate distances with a rotating disk velocimeter similar to that used in Ref. 1.

Results and Discussion

During preheating with the plasma gas, the substrate temperature varied with torch distance as shown in Fig. 2. Also shown on this figure are the back side substrate temperatures during spraying.

The average particle velocity data are shown in Fig. 3. Notably there is little difference in average particle velocity between 4 in. (240 ft/sec) and 7 in. (215 ft/sec).

The actual deposition patterns and cross-sectional views are shown in Fig. 4, 6, 8, and 9, and will be discussed in order of increasing torch-to-substrate distance. Before these cross sections are examined, it must be remembered that the particles were sparsely deposited on the substrate to permit easy examination of the particle configuration. For this reason, a cross section will not show a series of particles side by side, but only isolated particles. Also, the deposition efficiency decreases as torch-to-substrate distance increases, so that the 4-in. specimen shows several particles on top of the same spot while lower particle concentrations are seen at increasing torch-to-substrate distances.

4-in. torch-to-substrate distance. At a distance of 4 in. the plasticity of the

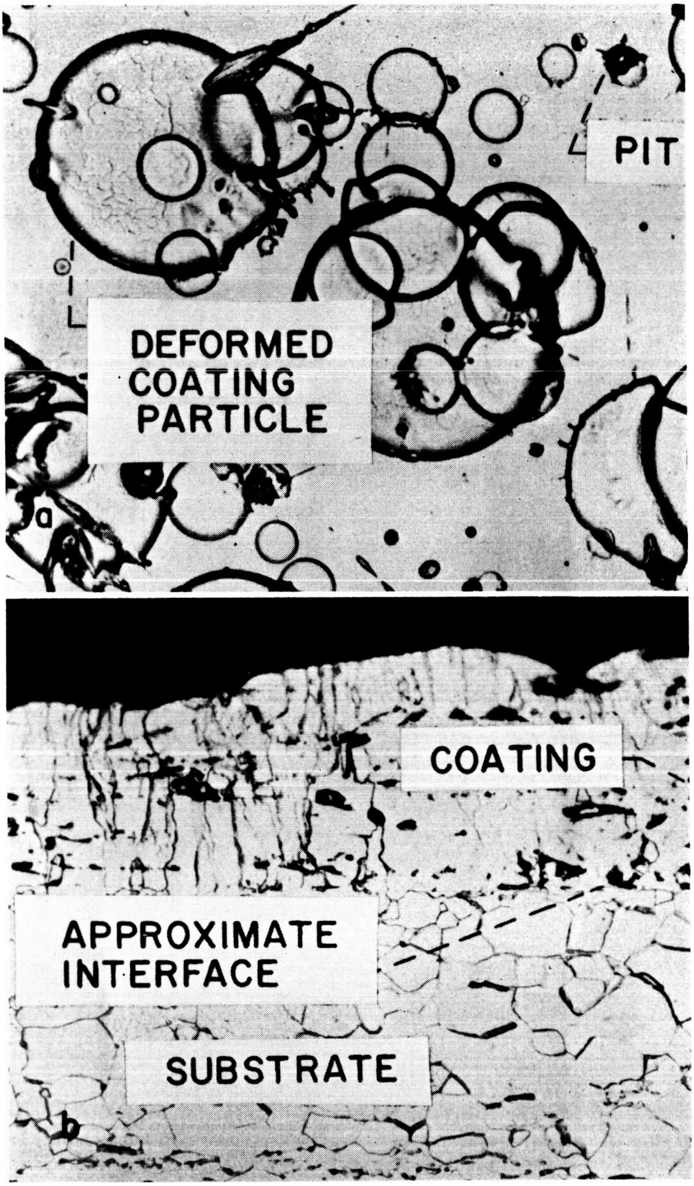

FIG. 4. As-sprayed and cross-section views for a 4-in. torch-to-substrate distance. Substrate temperature: ~2700 F.

particles is maximum for the range investigated. Most of the particles which struck the substrate were deformed into flat, almost circular disks and had sufficient plasticity to fold over previously deposited particles (Fig. 4a). Most of the particles

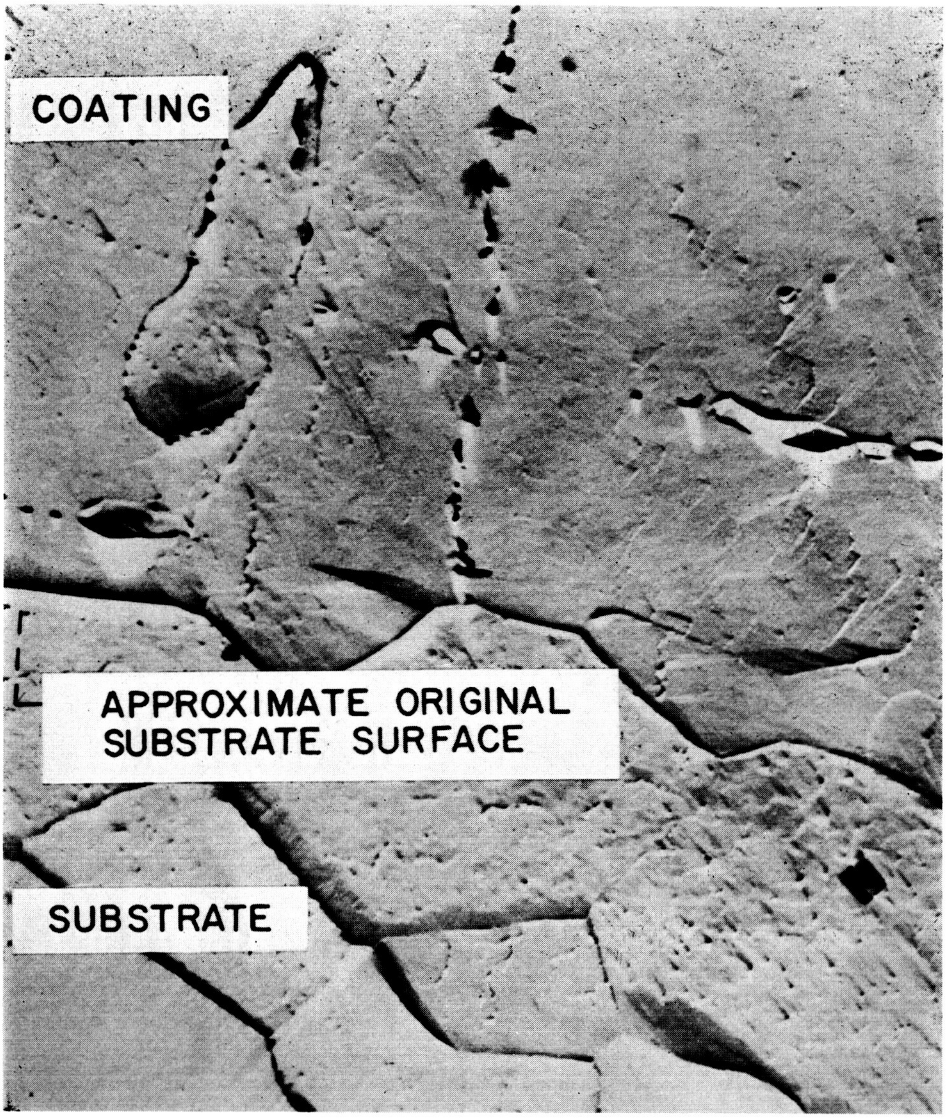

Fig. 5. Electron micrograph of a cross-section of the coating and substrate for a 4-in. torch-to-substrate distance. Etchant: Murakami's Reagent; 10,000 ×.

were, therefore, at high temperature as evidenced by the relatively few pits blasted into the substrate surface. The pits which are present were not filled by the flow of material from the particles next to them. This may indicate that dynamic resistance to flow (as governed by surface tension, velocity, temperature, etc.) is still sufficiently high to prevent such a discontinuous flow. This may well point to a

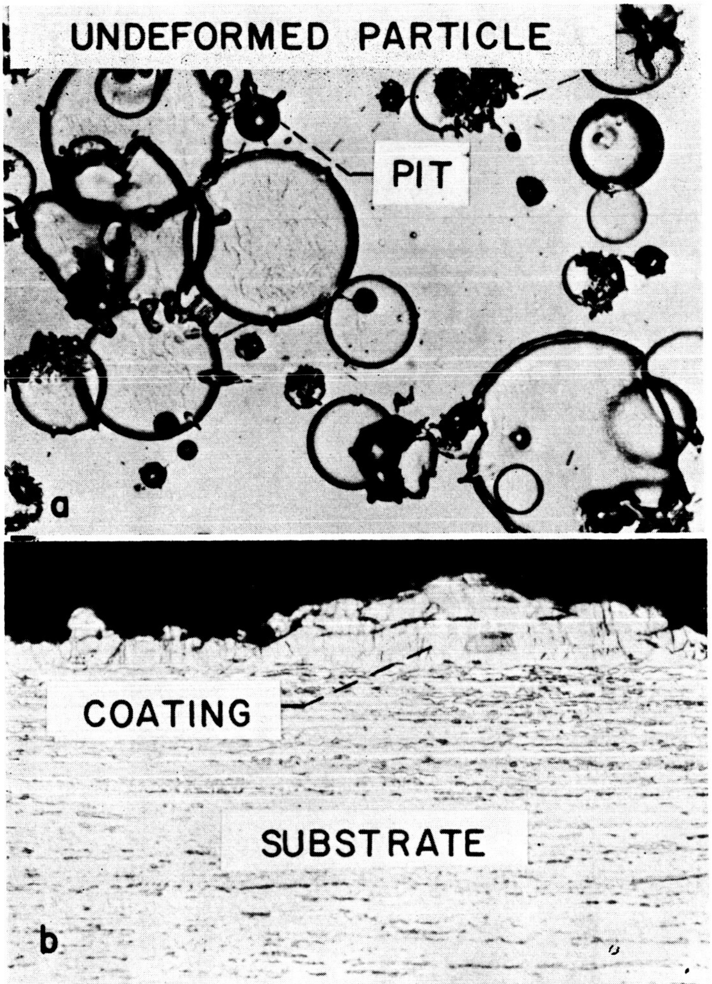

FIG. 6. As-sprayed and cross-section views for 5-in. torch-to-substrate distance. Substrate temperature: ~2400 F.

reason for difficulties when spraying on grit-blasted surfaces, where numerous surface discontinuities are present.

A cross-section view of the deposit and substrate (Fig. 4b) reveals large columnar grains perpendicular to the recrystallized structure of the substrate. The original interface has been almost completely eliminated. This appears to have occurred by each substrate surface grain growing into the coating. In some areas it is apparent that this growth continued even into the subsequent particles that landed on the first particle, i.e., the columnar grain boundaries are continuous through the rows of voids which mark interparticle boundaries. This continuity of columnar grains is better illustrated in Fig. 5, which

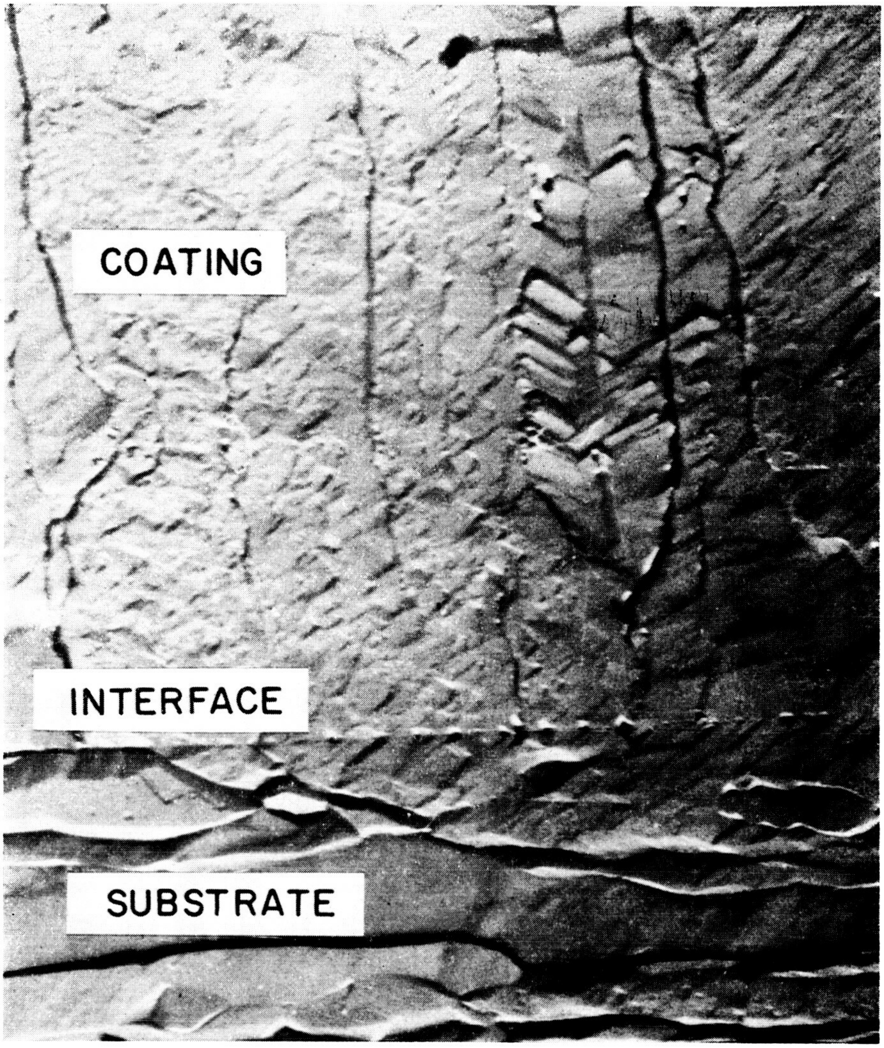

FIG. 7. Electron micrograph of a cross section of the coating and substrate for a 5-in. torch-to-substrate distance. Etchant: Murakami's Reagent; 10,000×.

is an electron micrograph. Here it can be seen that the columnar grains do grow through the original particle-to-particle interfaces, which appear as rows of voids in this figure. These voids are thought to be caused by surface oxides or absorbed gases. They should not be due to the presence of tungsten nitride since this compound is thermodynamically unstable at these temperatures as is discussed in Ref. 2.

5-IN. TORCH-TO-SUBSTRATE DISTANCE. In the 5-in. distance, the particles were similar to those at 4 in.; however, both the number and the size of the pits increased.

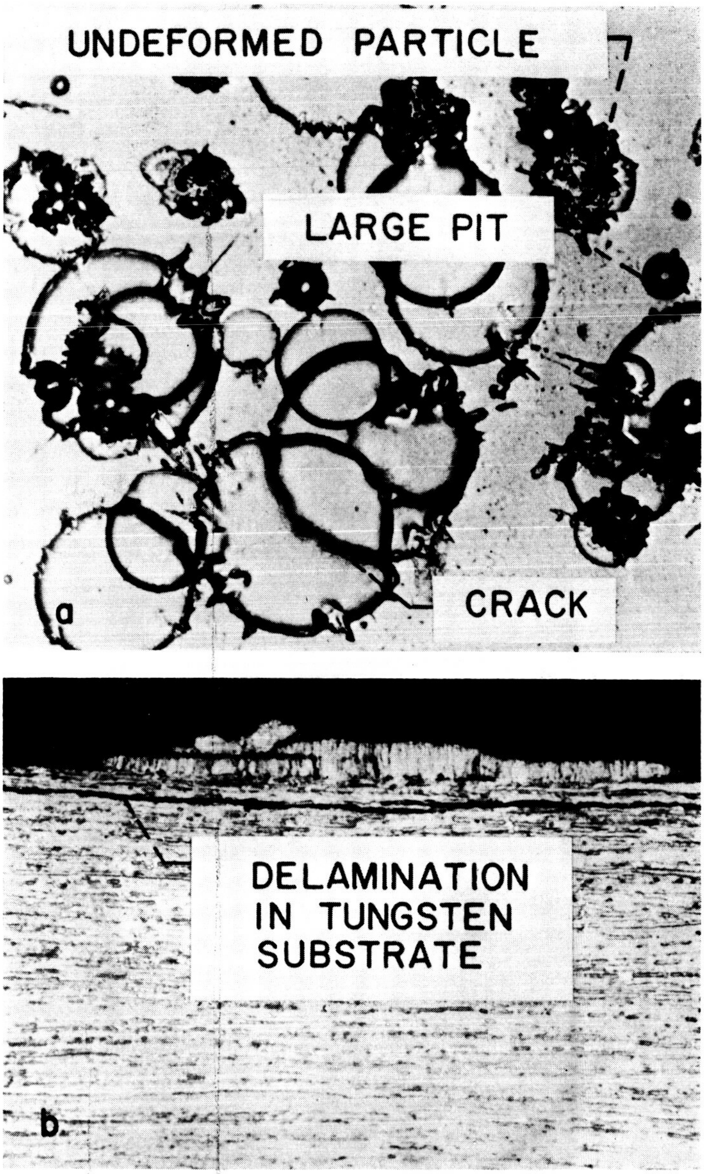

FIG. 8. As-sprayed and cross-section views for a 6-in. torch-to-substrate distance. Substrate temperature: ~2100 F.

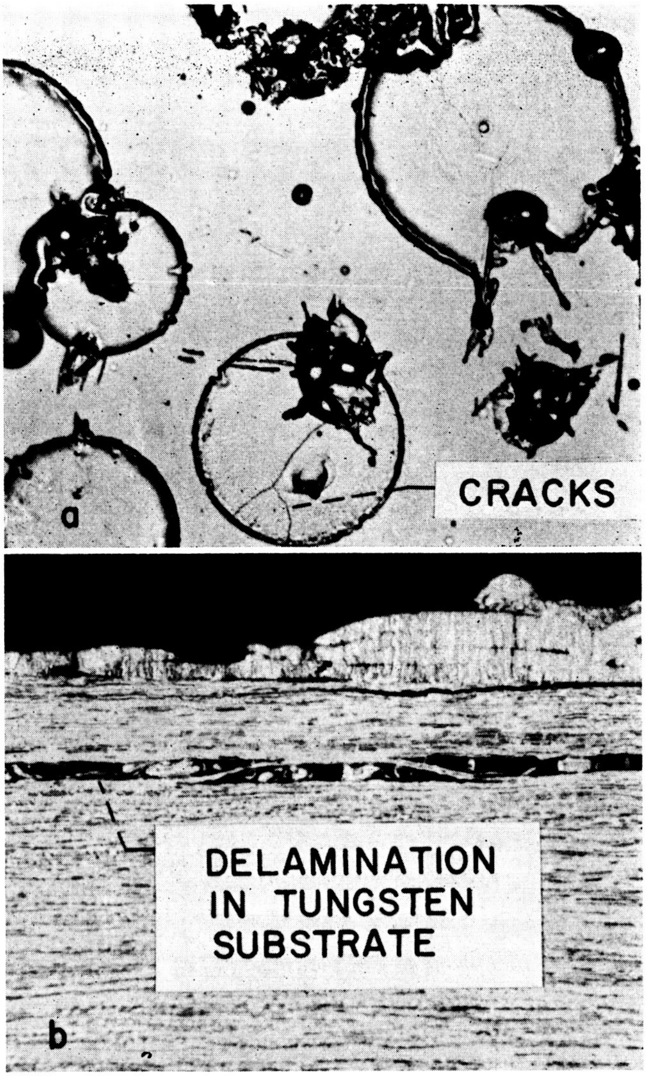

FIG. 9. As-sprayed and cross-section views for a 7-in. torch-to-substrate distance. Substrate temperature: $\sim$2050 F.

The amount of undeformed particles* had increased slightly (Fig. 6) (2).

* The undeformed particles were determined to be raised portions on the substrate by microscopic analysis of focus points.

The cross section (Fig. 6b) shows that the particle-to-substrate interface has been completely eliminated while maintaining an essentially "as-worked" structure in the substrate. Consistent with this, the columnar grains in the coating are smaller. Some evidence of particle-to-particle interfaces can be seen, but they are not sharp lines of demarcation. Figure 7 is an electron micrograph showing the coating and the substrate.

The original substrate surface is evidenced by the row of voids, but a metallurgical bond exists and there is continuity between grains of the substrate and of the coating. This situation exists even though there is essentially no recrystallization of the substrate. The relative size decrease in the columnar grains which occurred at this distance as compared to the size of the grains which occurred at 4 in. may be seen by comparing Fig. 5 and 7.

6 - IN. TORCH - TO - SUBSTRATE DISTANCE. The coupon surface (Fig. 8a) showed larger pits and a greater amount of undeformed particles than seen in Fig. 6a. The particles still exhibited a considerable amount of plasticity as can be seen by the amount of particle overlap. However, in spite of this plasticity, some particle cracking was observed.

The cross section (Fig. 8b) is similar to that obtained at 5 in. but the columnar grain structure is much finer. (The dark line is a delamination of the tungsten substrate.)

7 - IN. TORCH - TO - SUBSTRATE DISTANCE. The greater distance of 7 in. produced many undeformed particles (Fig. 9a), and most of those that did deform show cracks. The size of the pits is increasing but their number is decreasing slightly (as compared to Fig. 8a). The increase in the pit size and the decrease in number of pits indicate that only the larger particles have sufficient energy to cause pitting of the colder substrate at this distance.

The particle-to-substrate bond is not complete (Fig. 9b), but a metallurgical bond still exists over a great portion of the interface. The interparticle interfaces are more obvious and the columnar structure can still be seen.

Summary of Results

From an examination of the plasma-sprayed particles on preheated substrates over a substrate temperature range of 2050 to 2700 F, certain trends are observed:

1. The amount of undeformed particles increases slightly with increasing torch-to-substrate distance.
2. Cracks are present in the particles deposited at greater distances.
3. The number of pits is a minimum at the shortest distance investigated.
4. The pit size increases with increasing torch-to-substrate distance.

From examination of the coating substrate cross sections, certain trends can also be seen:

1. The columnar grain size of the coating decreases with increasing torch-to-substrate distance.
2. Up to a 6-in. torch-to-substrate distance, good metallurgical bonds exist between the coating and the substrate, and at 7 in. a metallurgical bond exists over a considerable portion of the interface.

Conclusions

1. Metallurgically bonded tungsten coatings can be achieved by plasma spraying on polished tungsten substrates, without prior roughening of the substrate surface, if the substrate is first heated in an inert atmosphere (the substrate temperatures investigated ranged from 2050 to 2700 F).

2. It is possible to deposit such coatings while retaining the high-strength properties of an as-worked substrate, i.e., a metallurgical bond can be achieved at a substrate temperature below the substrate recrystallization temperature.

3. The type of columnar grains which appear in all the cross sections indicates that the coating-substrate bond is the result of substrate surface grains growing into the coating.

4. Even under very short-time conditions, almost complete elimination of the original coating-to-substrate interface can be achieved. This indicates extremely rapid short-range diffusion occurred at this interface.

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